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Second-order optical nonlinearities in dilute melt proton exchange waveguides in *z*-cut $LiNbO_3$

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Planar optical waveguides with different refractive indices are made in *z*-cut $LiNbO_3$ with a dilute proton exchange method using a system of glycerol containing $KHSO_4$ and lithium benzoate. The optical second-order susceptibilities of these waveguides are measured by detecting the 266 nm reflected second-harmonic signal generated by a 532 nm beam directed onto the waveguide surface. It is found for this kind of waveguides that in the waveguide region all the second-order susceptibilities take values of at least 90% of the original $LiNbO_3$ values for refractive index changes less than ~0.013 at the 632.8 nm wavelength, whereas the susceptibilities are strongly reduced for larger index changes. © *1996 American Institute of Physics.* [S0003-6951(96)04142-3]

The exchange of lithium ions with protons is a well-known method for making optical waveguides in $LiNbO_3$.[1] One of the applications of the proton exchange (PE) method is for fabrication of channel waveguides where the large nonlinear properties of the $LiNbO_3$ crystal are used for frequency doubling of light propagating through the waveguide. In this way compact green/blue laser sources can be made by frequency doubling diode laser light available at longer wavelengths. Especially nonlinear waveguides in $LiNbO_3$, where the phase matching of the interacting beams is achieved by the quasi-phase-matching method, have received much attention the last years since the largest nonlinear second-order coefficient d_{33} of $LiNbO_3$ can be used for the second-harmonic (SH) generation.[2–4]

Several authors have investigated how the $LiNbO_3$ nonlinear properties are influenced by the PE process and the annealed PE (APE) process, where the local proton concentration is lowered by a postannealing treatment of the PE waveguide.[5–11] Publications on the subject report rather different results of the nonlinear properties of the PE and APE waveguides, which are apparently related to the difficult task of probing the nonlinear properties of a layer only few microns thick at the surface of the $LiNbO_3$ crystal. The method used by Laurell and co-workers[8] described below seems, however, to be a useful measurement technique. A laser beam with a wavelength of 532 nm is directed onto the surface of the waveguide layer on the $LiNbO_3$ crystal and the reflected SH signal (266 nm) generated by the nonlinearity of the layer is then detected. Since the 266 nm SH wavelength absorption depth is only about 0.05 μm (Ref. 8) it can be assumed that the reflected signal is due to the nonlinear optical properties of the top of the waveguide layer and not the bulk properties; hence, the theory of Bloembergen and Pershan,[12] describing the SH reflection from the interface between air and a nonlinear medium, applies to this case.

From the measurement of the reflected SH signal with the 532 nm probe beam, Laurell and co-workers[8] found a 30-fold reduction in the nonlinearity of a *z*-cut APE waveguide compared to pure $LiNbO_3$. This reduction was found even for postannealing times much longer than in the case of any practical APE waveguide design. Later Bortz and co-workers[11] applied the same technique to APE waveguides made in *x*-cut $LiNbO_3$ substrates. Using a special polishing technique the d_{33} nonlinear coefficient was probed through the whole APE waveguide region. It could be seen that the nonlinear properties of the original PE region were highly reduced even after a postannealing treatment over a relatively long time.

Based on these results of Bortz and co-workers[11] the fabrication of waveguides by the dilute melt method[13–15] seems to us to be the best method for making channel waveguides for frequency conversion. With an exchange melt containing both protons and lithium ions the waveguide refractive index—and hence the local proton concentration in the waveguide—can be controlled by the relative concentration of protons and lithium ions. In this way one avoids the step of first introducing severe changes of the original $LiNbO_3$ crystal from a high proton concentration and here-

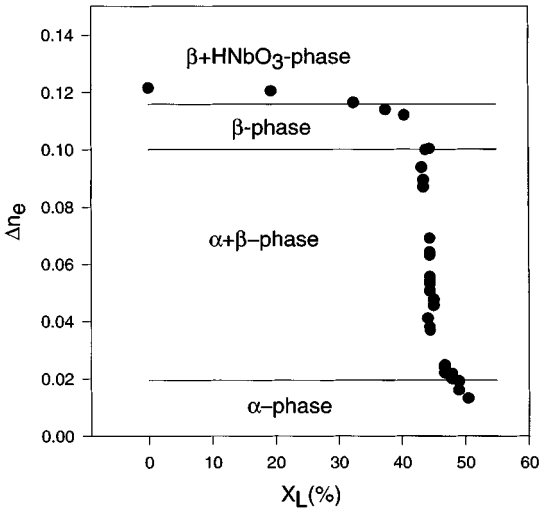

FIG. 1. Dependence of the change of the extraordinary refractive index on the exchange melt composition at 632.8 nm (taken from Ref. 16). The different crystal phase regions connected with the refractive index change are also shown.

TABLE I. Geometries used in the SH measurements. The orientations listed in column 2 are understood as follows: For example 70Xsp means that the incidence angle of the probe beam is 70°, the plane of incidence contains the $LiNbO_3$ crystal X-axis, and the incident light is s polarized while the p-polarized component of the SH light is detected.

Measurement No.	Orientation	Involved second-order susceptibilities	$X_L(0)$	α
1	45Ypp	$d_{22}, d_{15}, d_{31}, d_{33}$	50.6	0.18
2	45Ysp	d_{31}	50.4	0.17
3	70Ypp	$d_{22}, d_{15}, d_{31}, d_{33}$	50.9	0.16
4	70Ysp	d_{31}	50.2	0.17
5	70Xpp	d_{15}, d_{31}, d_{33}	51.2	0.16
6	70Xps	d_{22}	49.6	0.14
7	70Xss	d_{22}	48.6	0.15
8	70Xsp	d_{31}	50.4	0.17

after lowering the concentration by an annealing treatment. In order to achieve this, a new dilute melt (DM) method using glycerol containing $KHSO_4$ and lithium benzoate has recently been suggested.[16] Here we report on measurements of the reflected SH generation from a 532 nm laser beam incident at the surfaces of waveguides made in z-cut $LiNbO_3$ by this DM exchange method. To our knowledge, this is the first time a direct measurement of the nonlinear properties of DM waveguides is performed. The measurements are made on several waveguides fabricated with different exchange melt compositions. In this way the reduction in nonlinearities can directly be associated with the proton concentrations and, hence, the index changes introduced from the waveguide fabrication in $LiNbO_3$. This knowledge is important for the optimization of waveguide design in $LiNbO_3$ for frequency conversion.

Several waveguides were made with varying exchange time and varying exchange melt composition described via

$$X_L = 100\% \times \text{mol lithium benzoate} / (\text{mol lithium benzoate} + \text{mol } KHSO_4).$$

The amount of lithium benzoate determined by X_L was added to a solution of 2 g $KHSO_4$ dissolved in 200 ml glycerol. The exchange temperature was $T = 230$ °C. The dependence of the change of the extraordinary refractive index on the composition parameter X_L for waveguides made at

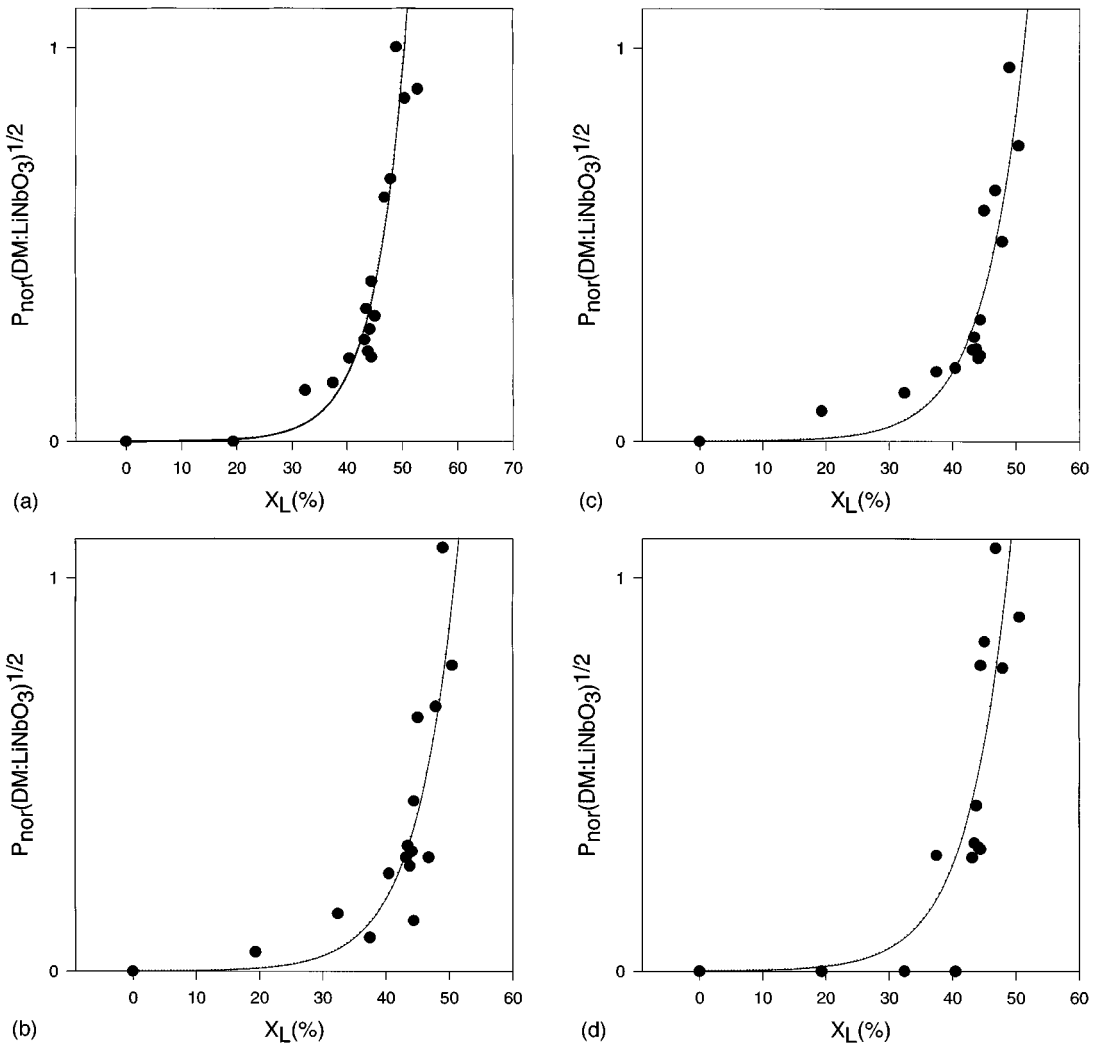

FIG. 2. Reflected SH signal (266 nm) from waveguides made with different DM compositions. The solid curves are the results of the fitting expression in Eq. (1). (a) Measurement No. 2 in Table I; (b) measurement No. 3 in Table I; (c) measurements No. 5 in Table I; (d) measurement No. 7 in Table I.

$T=230$ °C is taken from Ref. 16 and is shown in Fig. 1. The refractive index changes were determined at a wavelength of 632.8 nm.

A Q-switched frequency doubled Nd:YAG laser delivering 8-ns-long pulses at a repetition rate of 20 Hz was used for the SH measurements. The pulse energy was kept below 10 mJ and delivered on an area of about 1 cm^2 on the planar waveguide surface. After filtering with an interference filter, the SH light was detected by a photomultiplier tube connected to gated electronics. Eight different measurements on each waveguide were made. The geometry used in each measurement is described in Table I where also the involved nonlinear interaction coefficients are listed. For each geometry used, measurements with untreated $LiNbO_3$ substrates were also made as a reference.

Figure 2 shows how the measured SH signals vary with the waveguide fabrication parameter X_L. We have chosen to express the SH signal in terms of the quantity

$$P_{\mathrm{nor}}(\mathrm{DM{:}LiNbO_3})^{1/2} = P(\mathrm{DM{:}LiNbO_3})^{1/2}/P(\mathrm{LiNbO_3})^{1/2},$$

where $P(\mathrm{LiNbO_3})$ and $P(\mathrm{DM{:}LiNbO_3})$ denote the detected power of the reflected SH signal from a pure $LiNbO_3$ sample and from a DM $LiNbO_3$ sample, respectively. If the change in the $LiNbO_3$ linear optical properties introduced by the DM exchange process is neglected, the value of $P_{\mathrm{nor}}(\mathrm{DM{:}LiNbO_3})^{1/2}$ corresponds to the value of the effective nonlinear interaction coefficient $d_{\mathrm{eff}}(\mathrm{DM{:}LiNbO_3})$ normalized with respect to d_{eff} for pure $LiNbO_3$. In the following we consider only that part of the DM waveguides that has rather small induced index changes and proton concentrations, so it is reasonable to assume that the linear optical properties are nearly the same as in the case of untreated $LiNbO_3$.

The curves in Fig. 2 correspond to the geometries Nos. 2, 3, 5, and 7 listed in Table I. In all measurements listed in Table I, nearly the same behavior of the SH signal with X_L occurs as seen in Fig. 2. It seems reasonable to assume that $P_{\mathrm{nor}}^{1/2}(\mathrm{DM{:}LiNbO_3})$ can be related to X_L via an empirical expression of the form

$$P_{\mathrm{nor}}^{1/2}(\mathrm{DM{:}LiNbO_3}) = \exp\{\alpha[X_L - X_L(0)]\}, \tag{1}$$

when X_L takes values in the range $0 \leqslant X_L \leqslant X_L(0)$. The two symbols α and $X_L(0)$ are fitting parameters. The corresponding fitting curves from Eq. (1) are shown in Fig. 2. The fitting results are listed in Table I.

The average value of X_L, where the values of the effective second-order nonlinear coefficients are reduced to $1/e$ of the pure $LiNbO_3$ values, is $X_L(1/e)=44.0\%$. This value is obviously related to the steep line in Fig. 1, where a sudden change in refractive index change with X_L is seen. Hence, the dependence of the nonlinear properties on X_L is in agreement with the structural composition of proton-doped $LiNbO_3$ proposed by Rice.[17] According to Rice[17] the PE $LiNbO_3$ crystal consists of different crystal phases depending on the proton concentration. From the relation between proton concentration and change of the extraordinary refractive index taken from Ref. 16, these different phases are indicated in Fig. 1. Only in the pure α phase are no severe changes of the original crystal structure observed.[17] From comparison of Fig. 1 and the fitting results listed in Table I, it is concluded that for refractive index changes below $\sim$0.013 at 632.8 nm, all the nonlinear coefficients take values of at least 90% of the values of pure $LiNbO_3$.

In conclusion, we have investigated how optical nonlinearities in z-cut $LiNbO_3$ are influenced by the waveguide fabrication technique using a dilute melt proton exchange method. A relation between the values of the optical second-order susceptibilities and the exchange melt composition has been determined. Hence, information on how the second-order nonlinear properties of the $LiNbO_3$ crystal depend on the refractive index change is obtained. In this way we have found that for refractive index changes close to or less than $\sim$0.013 at 632.8 nm, all second-order nonlinear susceptibilities in the whole wave-guide region can be assumed to take at least 90% of the values of pure $LiNbO_3$. This is an important improvement of the fabrication of waveguides in $LiNbO_3$, for e.g., frequency conversion, whereas in the case of the commonly used annealed proton exchange waveguides, the nonlinear coefficients are not unaffected in the whole waveguide region.

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